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(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

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1. Your reference

GPS/BP6218572

- 2 APR 2004

2. Patent application number

(The Patent Office will fill this part in)

0407576.8

3. Full name, address and postcode of the or of each applicant (*underline all surnames*)
 IAN WALTERS
 BRUNEL HOUSE
 GORSEINON ROAD
 SWANSEA, SA4 9RU
 GB
Patents ADP number (*if you know it*)

8719353001

If the applicant is a corporate body, give the country/state of its incorporation

4. Title of the invention

USE OF RECYCLED RUBBER DUST IN VIRGIN RUBBER COMPOUNDS

5. Name of your agent (*if you have one*)
 "Address for service" in the United Kingdom to which all correspondence should be sent
(including the postcode)

 MEWBURN ELLIS LLP
 York House
 23 Kingsway
 London WC2B 6HP
Patents ADP number (*if you know it*)

109006

8836884001

6. Priority: Complete this section if you are declaring priority from one or more earlier patent applications, filed in the last 12 months.

Country

Priority application number
*(if you know it)*Date of filing
(day / month / year)

7. Divisionals, etc: Complete this section only if this application is a divisional application or resulted from an entitlement dispute (see note f)

Number of earlier UK application
(day / month / year)

8. Is a Patents Form 7/77 (Statement of inventorship and of right to grant of a patent) required in support of this request?

NO

Answer YES if:

- a) any applicant named in part 3 is not an inventor, or
- b) there is an inventor who is not named as an applicant, or
- c) any named applicant is a corporate body.

Otherwise answer NO (See note d)

Patents Form 1/77

9. Accompanying documents: A patent application must include a description of the invention.
Not counting duplicates, please enter the number of pages of each item accompanying this form:

Continuation sheets of this form	0
Description	11 ✓
Claim(s)	0 <i>[Signature]</i>
Abstract	0
Drawing(s)	0

10. If you are also filing any of the following, state how many against each item.

Priority documents	0
Translations of priority documents	0
Statement of inventorship and right to grant of a patent (Patents Form 7/77)	0
Request for a preliminary examination and search (Patents Form 9/77)	0
Request for a substantive examination (Patents Form 10/77)	0
Any other documents (please specify)	0

11. I/We request the grant of a patent on the basis of this application.

Signature(s)

M. Stoner *Agenda* Date 2 APRIL 2004

12. Name, daytime telephone number and e-mail address, if any, of person to contact in the United Kingdom

PATRICK STONER - 0117 926 6411
(patrick.stoner@mewburn.com)

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USE OF RECYCLED RUBBER DUST IN VIRGIN RUBBER COMPOUNDS**Introduction**

This disclosure is supplementary and clarificatory in
5 relation to my application 0322277.5 of 23 September 2003
and has to do with methods by which vulcanised waste
rubber can be recycled into new rubber compounds. The
information given specifically details formulations
developed as backing rubbers for mats, but the
10 formulating technique is also suited to other end
products. The formulations may require optimising for
particular manufacturing processes and product needs, but
this will not affect the principles of the development.
The products are an aspect of the disclosure.

15 The reworking of vulcanised waste rubber into new rubber
compounds is not a new concept, having been used within
the rubber industry for many years as a way to cheapen
compounds and in some instances to modify mould flow and
20 assist in the removal of air trapping during the moulding
process. However, due to the detrimental effect that
recycled rubber can have on both the processing
properties of uncured compound and the physical
properties of the final vulcanisate, the practice has
25 been limited. The ease of addition and the quantity of
recycled rubber added to a rubber compound is largely
dictated by its particle size, where a greater volume of
recycled rubber with larger particles can be added than

of small. It is also found that the incorporation of large cured rubber particles adversely affects the physical properties of a rubber compound, in particular tensile strength, tear, elongation at break and abrasion resistance. Conversely, the use of small particle sized rubber dust (<0.4mm) reduces the detrimental affect on physical properties, but is very much more difficult to add in large quantities without sacrificing the processing properties of the mix, e.g. handling during mixing, extrusion and calendering, due to a reduction in tack and uncured rubber green strength. An increase in compound viscosity may also be seen, leading to reduced moulding flow. Conventionally, the use of rubber crumb / dust as an additive in rubber compounds is limited to levels of 5% to 15% for products where the retention of tensile, tear, abrasion strength and compound processing is important. Greater addition levels are sometimes used, but for applications where lower physical and dynamic properties are acceptable. The reduction of the physical strength of a rubber compound when rubber dust is added can be attributed to;

- The rubber particles acting as localised stress raisers leading to the possible initiation of tear.
- The relatively low adhesive strength (when compared to the cohesive strength of the virgin

rubber) between the dust particles and the virgin compound giving a route for tear propagation.

It is considered likely that the process of grinding
5 rubber into a dust produces rubber particles that exhibit a level of surface activity due to a "de-linking" of sulphur-carbon cross-links. Under suitable vulcanisation conditions, recombination may occur, chemically bonding the dust particles to the surrounding virgin compound
10 matrix. If this is correct, the smaller the particle size of the rubber dust, the greater will be the surface area for a given rubber dust weight, and therefore the greater the opportunity for recombination to take place. It would therefore be preferable to use the smallest
15 particle sized rubber dust available within practical and commercial constraints to achieve the best physical properties for the vulcanised product. The addition of fine particle sized rubber dust (<40 mesh) is made progressively more difficult as the particle size reduces
20 and the resulting surface area increases. As with all fillers, full wetting of the virgin rubber compound matrix onto the surface of the dust particles is necessary to maintain compound homogeneity. The greater the surface area of the dust particles, the more
25 difficult it will be to achieve adequate levels of homogeneity. The ability of a compound to achieve good wetting of its ingredients will be affected by its viscosity, which in turn is affected by the average

molecular weight and the molecular weight distribution of the base polymer, and the use of plasticisers and processing aids. Inadequate wetting by the surrounding virgin compound will result in a dry crumbly mix with poor green strength and process flow.

5

Conventionally, processing oils, and other (soap based) processing aids are used to reduce compound viscosity, thereby assisting the wetting process, but when used in large amounts to assist the incorporation of high levels of rubber dust, they do little to retain either the processing characteristics of the compound or its cured physical properties. It is thought that the following may occur;

10 15

1. High additions of process oils, selected by solubility parameter for compatibility with the base polymer, may not be fully taken up by the uncured portion of the mix, causing some swelling of the rubber dust particles and further increasing dust particle surface area.

20

2. Soap based process aids have poor compatibility with the base compound which allows them to behave as internal lubricants. It is possible that they will coat the surface of the rubber dust particles, reducing the opportunity for either physical or chemical bonds to form between the dust particles and the virgin rubber matrix, leading to poor cured physical properties.

25

3. Neither the process oils nor the soap based process aids have the ability to cross link within the rubber matrix, and will have no beneficial affect on the cohesive strength of the uncured compound, but will reduce the cured modulus.

While the use of processing oil at relatively low levels that could be easily taken up by the base compound was found to be useful, successfully incorporating higher than normal levels of fine particle sized rubber dust depended upon identifying process aids that would not exhibit the characteristics listed above. Extensive laboratory studies identified liquid rubbers (very low molecular weight rubbers) to be effective in satisfying this requirement.

Liquid Rubbers

Very low molecular weight (liquid) rubbers are commercially available in a range of polymer types and viscosities. Some use of these materials has been made within the rubber industry as viscosity modifiers, but more often they are compounded for pressure sensitive adhesives, castable products, sealants and asphalt modifiers. Unlike rubber latices, liquid rubbers are not emulsions and can be added directly to rubber mixes within the normal rubber mixing process. As with all rubber blends, it is important to select a liquid rubber

that is compatible with the base polymer of the mix in terms of both solubility and cure performance. Our development work used compatible liquid rubber and base rubber types: liquid natural rubber with a NR/SBR base compound and a liquid nitrile rubber with a NBR base compound. The perceived advantages in using liquid polymers in combination with fine particle sized rubber dust can be listed as follows;

1. Liquid rubbers are fully capable of vulcanisation by conventional rubber cure systems and will therefore vulcanise within the base rubber matrix.
2. Although they are of low molecular weight (by polymeric standards), liquid rubbers are still of sufficiently high molecular weight not to cause swelling of rubber dust particles, but will assist in achieving good surface wetting.
3. Liquid rubbers should have no contaminating effect on the surface activity of the rubber dust particles, and therefore should have no inhibiting effect on the formation of crosslinks with the base rubber matrix.
4. Liquid rubbers will allow good adhesion between the rubber dust particles and the base rubber matrix, leading to improved processability.
5. Lower compound viscosities are achieved for improved mould flow.
6. Being fully compatible with the base polymer of the compound, liquid polymers will not bloom to the

surface of the rubber, offering improved adhesion to bonded substrates.

US 4481335 proposed the use of liquid rubber containing its own curing agent to treat crumb rubber, creating a crumb rubber preparation with a softened surface which can be directly compression moulded. Alternatively, it was described to incorporate the pretreated crumb rubber into rubber base compounds as an extender or filler.

In the present proposals, by contrast, the liquid rubber is used more in the fashion of a processing aid i.e. as a mix component rather than in a pretreatment. It is used in relatively more substantial quantities, constituting a genuine proportion of the eventual rubber compound and at least partially replacing other kinds of processing aids.

The addition level of liquid rubber to a mix is dependent upon the amount and particle size of the rubber dust used within the formulation, the use of other fillers (reinforcing and inert), and the viscosity of the base polymer within the mix. Pre-mastication of a natural rubber base polymer may be beneficial in achieving subsequent wetting out of the rubber dust, and may allow a reduced addition level of the liquid polymer to achieve the necessary viscosity. Similarly, selection of low viscosity grades of synthetic polymers may be beneficial for the same reason.

Formulation and Mixing Details

The formulations given below show a NR/SBR compound and a NBR compound, both containing just over 40% by weight of rubber dust. Each formulation has been successfully mixed and cured to give matting products with satisfactory properties, although neither compound has been optimised for specific products or processes. Any changes to the formulations to achieve compound optimisation, including the grade and quantity of the liquid rubber used, are quite acceptable, providing the changes do not reduce the effectiveness of the liquid polymer within the system. Similarly, the mixing method employed suited the equipment available for the development work, and should not be considered prescriptive. The fill factor and addition times will need to be established to suit the machine in which the compound is mixed, although it is recommended that the dump temperature does not exceed 110°C so that an adequate scorch safety can be maintained. While an internal mixer with tangential rotors was used for the development mixing, it is anticipated that the use of a mixer with intermeshing rotors will also give satisfactory results.

Natural Rubber/SBR

Ingredients	phr	
Pre-masticated NR	32.250	100phr NR/7.5phr Pepton 66. ML1+4@100°C 30 tot 55
DRP 40	10.000	Liquid NR Supplied by Elementis
SBR 1502	60.000	
N339	40.000	
Aromatic Process Oil	40.000	
Rubber Dust	135.000	60 mesh from ground tyre tread
Zinc Oxide	4.000	
Stearic acid	2.000	
Antidegradent BPH	1.500	Non-staining (Vulkanox BKF)
Antidegradent MMBI	1.000	Non-staining (Vulcanox ZMB2/C5)
TMTD - 80%	0.250	
TBBS	1.300	
Soluble Sulphur	2.750	
TOTAL		330.050

S.G. 1.120

Hardness 50 IRHD

5 Mixed using a 2.6 litre capacity laboratory Banbury mixer with tangential rotor mixing action.

Fill Factor 95%

	Mixing Cycle:	(Time from Start)
	Natural Rubber/SBR/DPR40	0 minutes
10	Black/Oil/Rubber Dust	0.5 minutes
	Zinc Oxide/Stearic Acid	1.0 minutes
	Antidegradents/Cure System	1.5 minutes

Dump to temperature @ 110°C

15

MDR @ 180°C

ML 2.22

ts2 0.77

t50 0.81

20

t90 1.44

MH 6.81

Nitrile

Ingredients	Phr	
NBR 34.50	80.000	
Nipol 1312 LV	20.000	(Supplied by Zeon)
N660	40.000	
Rubber Dust	110.000	60 mesh from ground W.O.M. mat edge trimmings
D.O.P.	10.000	
Zinc Oxide	5.000	
Stearic Acid	1.000	
A.D.P.A.	2.000	(Permanax BLW)
CBS	2.000	
TMTM	0.500	
MC Sulphur	2.000	
TOTAL		272.500

S.G. 1.150
 Hardness 50 IRHD

5 Mixed using a 2.6 litre capacity laboratory Banbury mixer with tangential rotor mixing action.

Fill Factor 95%

	Mixing Cycle:	(Time from Start)
	NBR/Nipol 1312 LV	0 minutes
10	Black/Oil/Rubber Dust/Sulphur	0.5 minutes
	Zinc Oxide/Stearic Acid	1.0 minutes
	Antidegradents/Accelerators	1.5 minutes

Dump to temperature @ 110°C

15 MDR @ 180°C
 ML 1.26
 ts2 0.50
 t50 0.54
 20 t90 0.76
 MH 8.17

Summary

25 The development described has shown that with the use of liquid polymers it is possible to incorporate higher than normal levels of small particle sized rubber dust into new rubber mixes while retaining reasonable processing characteristics and useful vulcanisate properties. The
 30 use of small particle rubber dust is seen as important for the retention of cured physical properties, while the use of liquid rubbers allows improved incorporation of the dust into the mix through enhanced surface wetting of the rubber dust particles. Preferably the particles will
 35 pass a No. 40 mesh, more preferably a 60 mesh or 80 mesh or smaller. Alternatively stated all or substantially all the particles are 0.25mm or smaller, preferably 0.2mm

or smaller, more preferably 0.15mm or smaller.

Remarkably, the present technology enables take-up into a strong, uniform rubber composition of at least 30, 40, 50, or even 60 wt% or more of such dust into the compound

5 leading to a high quality cured product. This development has the potential for significantly reducing disposal costs of cured rubber waste by recycling it into new products, and offers opportunities for cheaper compounds, both through lower material costs and by
10 improved weight to volume ratios.

Further improvements can be made by optimising the combination of the rubber dust particle size and the with the use of liquid rubbers. These formulations are
15 suitable for roll-processing to make mats or flooring articles as well as a variety of other products.

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